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THE DISTRIBUTION AND ABUNDANCE OF GAMMA EMITTING RADIONUCLIDES IN LAKE ONTARIO SEDIMENTS

Report No. 85-3-K

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ABSTRACT

The distribution of gamma emitting radionuclides in Lake Ontario sediments was investigated. Samples were collected using a systematic design in the vicinity of Pickering and Darlington, and supplemented by lakewide offshore samples. Naturally occurring ^{40}K was the predominant source of gamma activity. ^{60}Co was the only potentially CANDU released radionuclide which showed a distributional association with the Pickering 'A' NGS discharge.

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EXECUTIVE SUMMARY

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The distribution of gamma emitting radionuclides in Lake Ontario, was determined from sediments samples. Eleven gamma emitting radionuclides were detected in samples of Lake Ontario sediments. Naturally occurring radionuclides, which were dominated by the ubiquitous ^{40}K, accounted for the bulk of gamma activity. Lakewide gamma activity appeared to increase with sediment percent organic matter and depth. The most probable indicators of recent CANDU activity were ^{60}Co and ^{134}Cs, which collectively accounted for much less than 1% of total gamma activity. Detectable ^{60}CO was distributed primarily within a 2 km radius of the Pickering 'A' NGS CCW discharge. Highest ^{60}Co activities were observed from the shallower locations most proximal to the discharge channel. A lone occurrence of ^{134}Cs was detected at 22 km east of the Pickering site. The source of sporadic occurrences of ^{60}Co and ^{134}Cs at 12 to 22 km east of the Pickering site was not apparent. ^{60}Co activities observed in the lake basins probably originated from the Niagara River watershed. ^{137}Cs activities in the vicinity of Pickering and Darlington were not unique, and therefore probably originated from weapons testing.

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TABLE OF CONTENTS

	Page
INTRODUCTION	1
METHODS AND MATERIALS	1
Data Analysis	3
RESULTS	3
DISCUSSION	10
CONCLUSIONS	11
REFERENCES	13
DISTRIBUTION	-last page-

LIST OF TABLES

		Page
1.	Mean Gamma Radionuclide Activity In Lake Ontario Sediments	4
2.	Multiple Correlation Matrix For Gamma Emitting Radionuclides And Total Gamma Activity	6
3.	Stepwise Multiple Regression Results a) Lakewide Total Gamma Activity b) ^{137}Cs Activity In The Vicinity Of Pickering And Darlington	6
4.	Gamma Radionuclide Activity In Lake Ontario Basins Attributable To Radionuclides Of Potential CANDU Origin	8

LIST OF FIGURES

		Page
1.	Lake Ontario Sediment Sampling Locations a) Nearshore b) Lakewide (Depths Bracketed)	2
2.	Distribution Of Cesium-137 In The Nearshore Sediments (3, 6, 12 and 20 m Depths) From Pickering To Darlington Transects Initiated From 4 km West Of Pickering 'A' NGS Discharge (Transect 3) To 4 km East Of Darlington GS (Transect 16)	7
3.	Distribution Of Cobalt-60 In The Nearshore Sediments (3, 6, 12 And 20 m Depths) From Pickering To Darlington Transects Initiated From 4 km West Of Pickering 'A' NGS Discharge (Transect 3) To 4 km East Of Darlington GS (Transect 16)	9

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To Mr. F.J. Kee
Director of Research

DISTRIBUTION AND ABUNDANCE OF GAMMA EMITTING RADIONUCLIDES IN LAKE ONTARIO SEDIMENTS

INTRODUCTION

It is generally reported in the literature that sediments can effectively adsorb gamma radionuclides in the aquatic environment (Aston and Stanners 1982ab, Stanners and Aston 1982). The equilibrium distribution coefficient (Kd) between water and sediments is reported for many of the gamma emitting radionuclides to be greater than 10^3 (Booth 1975). Because of this strong affinity for sediments, exchange between water and sediments is likely to dominate the environmental dynamics of many gamma radionuclides (Ontario Hydro Research Division (OHRD) Report 82-271-K).

The objective of this study was to determine abundance and distribution in Lake Ontario of gamma emitting radionuclides by examining both inshore and deep lake basin areas. Factors that may explain their distribution in the sediment were also examined. The present study is part of a larger research initiative to determine the behavior of gamma emitting radionuclides released to the aquatic environment.

Routine environmental and radiological dose assessments of gamma radionuclides released in the vicinity of Pickering 'A' NGS are contained in reports prepared by the Health and Safety Division for RMEP (SSD-AR-79, SSD-AR-80-1, SSD-AR-82-1). These reports demonstrate that radiological doses resulting from gamma radionuclides are very low, and a small percentage of natural background radioactivity.

METHODS AND MATERIALS

The inshore distribution of gamma emitting radionuclides was determined in an area bordered by the Pickering 'A' NGS in the west and the site of the Darlington NGS in the east and the 20 m depth contour of Lake Ontario (Figure 1b). A systematic sampling design was employed. Sampling locations were arranged in a 18 transect X 4 depth grid. Transects, perpendicular to shore, were two kilometers apart, initiating from four kilometers west of Pickering 'A' NGS and

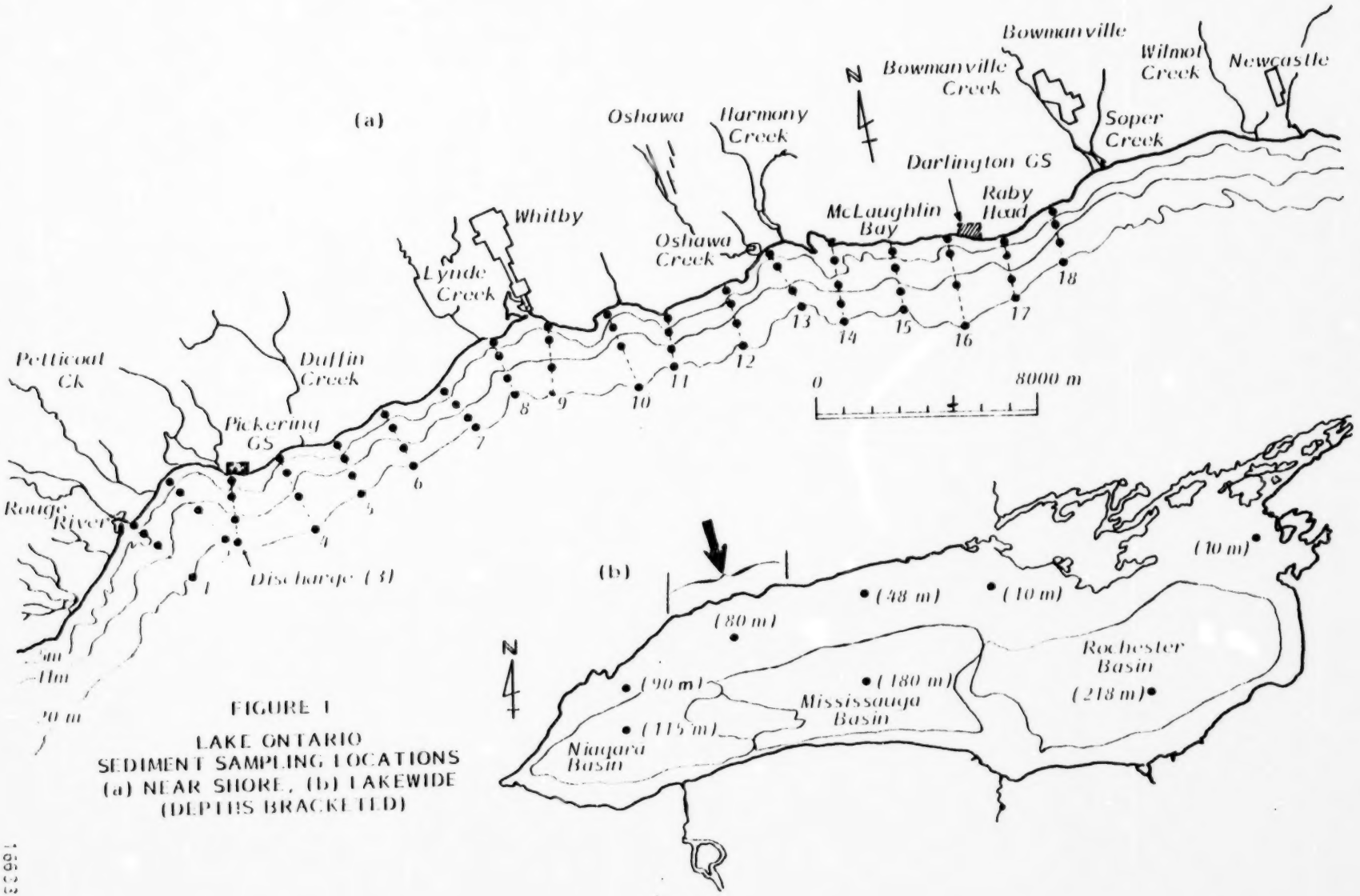

FIGURE 1

LAKE ONTARIO
SEDIMENT SAMPLING LOCATIONS
(a) NEAR SHORE, (b) LAKEWIDE
(DEPTHS BRACKETED)

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extending four kilometers east of the Darlington NGS site. Duplicate (one to be analyzed/one to be frozen) samples were collected at the 3, 6, 12, and 20 m depths of each transect. Additionally samples were taken throughout Lake Ontario at pre-selected locations (Figure 1b), courtesy of Dr. R.L. Thomas (CCIW).

Sediment samples were collected using a ponar grab. Samples were dried at 60^{0} C to constant weight and after gentle disaggregation placed into beakers for analysis. Weighed samples were then counted for a 60 h period with a Ge (Li) Canberra detector (14.3% efficiency) by the Analytical Services Section. Gamma radionuclide activities were determined using a computerized program (Scorpio-POP 105) which analyzed the pulse height spectrum of each radionuclide. All results were expressed as Bq/kg dry weight of sample.

Two physical characteristics of the sediment were measured, particle size and percent organic content. These have been shown to influence the variability in distribution of gamma emitting radionuclides in the sediment (Aston and Stanners 1982b). Particle size was determined by the wet sieve method (Folk 1968). Organic content of the samples was determined by loss on ignition at 550^{o} C.

Data Analysis

A multiple correlation matrix was inspected to determine whether gamma radionuclides were co-distributed. Associations with physical parameters were determined from the lakewide distribution of total gamma activity subjected to stepwise multiple regression against the measured variables (depth and percent organic).

Radionuclides potentially of CANDU or other nuclear sources (eg at Rochester, Scriba) origin (^{137}Cs, ^{134}Cs, ^{60}Co), were analyzed separately for samples in the vicinity of Pickering and Darlington. Gamma activity relative to shoreline location and depth was plotted, and inspected for any relationships with the measured physical variables. Stepwise multiple regression was used to determine whether measured physical parameters accounted for the distribution. Additionally, samples taken from the in-shore zone of 48-90 m, and samples taken from the lake basins were inspected for the presence of radionuclides.

RESULTS

A total of eleven detectable gamma emitting radionuclides were present in sediment samples collected from the inshore zone and basins of Lake Ontario (Table 1). Total gamma activity averaged

TABLE 1

MEAN GAMMA RADIONUCLIDE ACTIVITY
IN LAKE ONTARIO SUBSTRATES

Gamma Emitting Radionuclide[1]	Inshore Zone Depths 3-21m		Inshore Zone Depths 40-90m		Lake Basins Depths 115-220m	
	Bq/kg	Percent	Bq/kg	Percent	Bq/kg	Percent
^{40}K	391.	77.9	419.	70.8	850.	52.7
^{228}Ac	10.8	2.1	17.4	2.9	45.6	2.8
^{212}Pb	15.7	3.1	24.4	4.1	67.8	4.2
^{212}Bi	6.4	1.3	10.8	1.8	28.0	1.7
^{208}Tl	3.9	0.8	6.2	1.0	15.7	1.0
^{226}Ra	38.7	7.7	62.4	10.5	163.	10.1
^{214}Pb	14.1	2.8	21.3	3.6	64.1	4.0
^{214}Bi	15.3	3.1	23.2	3.9	61.4	3.7
^{137}Cs	5.5	1.1	7.2	1.2	319.	19.1
^{60}Co	0.5	0.1	*		1.5	0.1
^{134}Cs	0.1	0.0	*		*	
Total	502.		592.		1616.	
Mean Depth (m)	8.4		72.7		171.0	
Percent Organic	1.1		0.9		11.5	
Number of Samples	51		3		3	

* not detected

1 Origin

Natural; ^{40}K, ^{228}Ac, ^{212}Pb, ^{212}Bi, ^{208}Tl, ^{226}Ra, ^{214}Pb, ^{214}Bi
Nuclear Facilities; ^{60}Co, ^{134}Cs, ^{137}Cs
Weapons Testing; ^{137}Cs, half life of 30.2 years
^{134}Cs, half life of 2.05 years

502 Bq/kg and 592 Bq/kg for two depth ranges of the inshore zone, and 1616 Bq/kg for samples collected from the lake basins. Generally the level of gamma activity appeared to increase with depth for all detectable radionuclides, however total radioactivity present in the lake basins was in excess of three times the average calculated for either inshore depth range. The distribution of gamma emitting radionuclides representing both natural background and long-lived weapons testing radionuclides were strongly correlated with total radioactivity as shown by correlation coefficients ranging from r=0.88 to r=0.94 (Table 2). Total gamma activity was subjected to stepwise multiple regression against the variables depth and organic content. Depth was highly correlated with organic content (r=0.85) and was eliminated from the analysis. Organic content accounted for 83% of the variability in the data set (r=0.91) (Table 3). Three points were poorly predicted by the relationship with organic matter. Poorest fit was observed for samples from the deep water sites at 115 m and 180 m and the 20 m depth site located just off the shore from the town of Ajax (transect 7). Higher than expected levels observed in the Ajax area are unexplained.

Of the eleven gamma emitting radionuclides found, only three were potentially of CANDU origin - ^{134}Cs, $^{137}137$, and ^{60}Co. Of the three, the two more probable indicators of recent CANDU activity were ^{134}Cs and ^{60}Co, which were poorly correlated with other radionuclides and total radioactivity (Table 2). Together these two radionuclides accounted for less than one percent of the total inshore and offshore gamma activity (which consisted primarily of ^{40}K, a naturally occurring radionuclide). The uncertainty attributed to ^{137}Cs as an indicator of CANDU activity is due to the fact that environmental ^{137}Cs will originate both from weapons testing and from the operation of nuclear facilities.

The presence of detectable ^{134}Cs and ^{137}Cs in the inshore sampling grid, as well as throughout the lake differed markedly. Detectable ^{134}Cs was only observed from a single sample collected twenty-two kilometers east of the Pickering 'A' NGS discharge (transect 14 - McLaughlin Bay). ^{137}Cs activity was detected in all samples collected throughout the lake. The occurrence of ^{137}Cs was not unique at the immediate vicinity of the Pickering 'A' NGS, but occurred at generally similar levels throughout the inshore zone (Figure 2). A stepwise multiple regression indicated, that in the vicinity of Pickering and Darlington depth and percent organic content accounted for 42% (r=0.65) of the distribution of ^{137}Cs (Table 3). A single occurrence of 36.3 Bq/kg at transect 7-20 m was poorly predicted by the regression variables. Deletion of this point from the data set resulted in re-ordering the physical variables and improving the regression (r=0.88).

TABLE 2

MULTIPLE CORRELATION MATRIX FOR GAMMA EMITTING RADIONUCLIDES AND TOTAL GAMMA ACTIVITY

	^{228}Ac	^{212}Pb	^{212}Bi	^{208}Tl	^{208}Ra	^{214}Pb	^{214}Bi	^{60}Co	^{137}Cs	^{134}Cs	TOTAL
^{40}K	.00	.80	.80	.79	.74	.71	.70	.14	.76	-.00	.94
^{228}Ac		.99	.99	.98	.98	.96	.96	.06	.80	-.10	.94
^{212}Pb			.99	.98	.98	.95	.96	.09	.80	-.13	.94
^{212}Bi				.98	.97	.94	.95	.06	.80	-.12	.94
^{208}Tl					.96	.93	.94	.08	.87	-.10	.93
^{208}Ra						.98	.99	.06	.88	-.10	.91
^{214}Pb							.98	.08	.88	-.06	.89
^{214}Bi								.02	.85	-.07	.88
^{60}Co									.11	-.03	.13
^{137}Cs										-.04	.92
^{134}Cs											-.00

TABLE 3

STEPWISE MULTIPLE REGRESSION RESULTS

a) Lakewide Total Gamma Activity

Variable	Coefficient	Standard Error	Constant	Multiple R Squared	Correlation Coefficient
% Organic	101.406	6.209	401.665	0.83	0.91

b) ^{137}Cs Activity in the Vicinity of Pickering and Darlington

	Variable	Coefficient	Standard Error	Constant	Multiple R Squared	Correlation Coefficient
A-	Depth	0.529	0.142		0.26	0.51
	% Organic	3.063	0.347	-2.112	0.42	0.65
B-	% Organic	4.058	0.361		0.75	0.86
	Depth	0.155	0.065	-0.848	0.78	0.88

A= All samples collected in the inshore vicinity of Pickering and Darlington.

B= Samples in the Pickering and Darlington vicinity with the transect 7 20 m sample deleted.

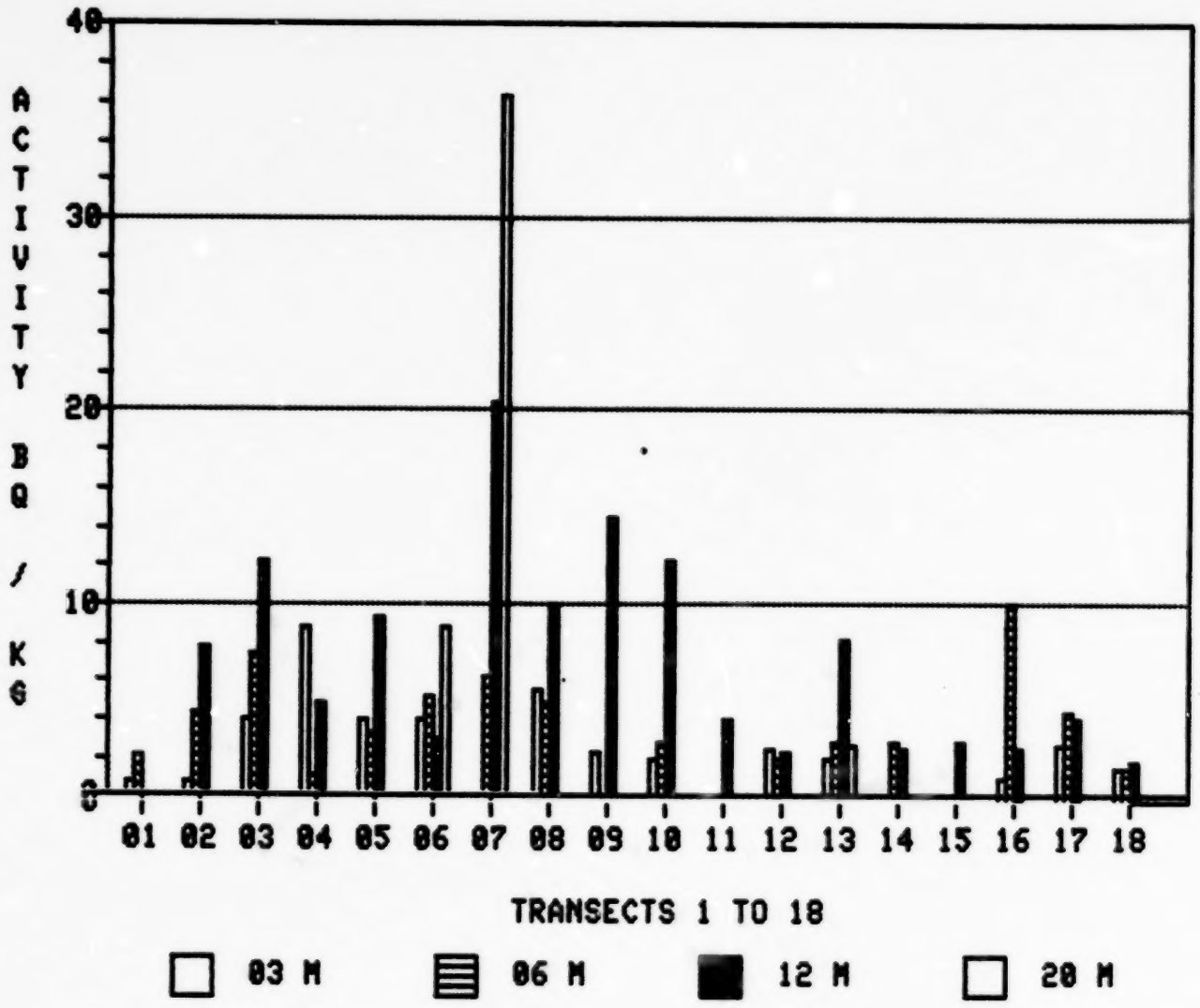

FIGURE 2

Distribution of Cesium-137 in the Nearshore Sediments (3,6,12 and 20 M Depths) from Pickering to Darlington. Transects Initiated from 4 km West of Pickering 'A' NGS Discharge (Transect 3) to 4 km East of Darlington GS (Transect 16).

Detectable ^{60}Co was observed in nine samples from the inshore zone however it only occurred at the 3 and 6 m depths in the vicinity of the Pickering 'A' NGS discharge (Figure 3). A peak concentration of 17.8 Bq/kg was observed from the 3 m discharge location. At locations east of the Pickering vicinity ^{60}Co was detected in samples from the 12 m depth only. Occurrences at the 12 m depth along transects 9-10 and 13-14, were observed at 12-14 and 20-22 km east of the Pickering 'A' NGS CCW discharge. The distribution of ^{60}Co was related only to depth and distance from Pickering 'A' NGS discharge.

Samples collected offshore from Toronto (90 m), Darlington (80 m), and Port Cobourg (48 m), were devoid of detectable ^{134}Cs and ^{60}Co (Table 4). ^{137}Cs was, however present at levels generally in excess of activities observed at the shallower depths. Samples collected from the Niagara Basin (115 m), Missisauga Basin (180 m), and Rochester Basin (218 m) were devoid of detectable ^{134}Cs, however ^{60}Co and ^{137}Cs were present. ^{60}Co levels of 0.78-2.55 Bq2/kg were present in the basins, and were generally not higher than activities observed in the Pickering and Darlington area. ^{137}Cs levels however averaged 58 and 48 times larger than those levels observed at the inshore depths of 3-21 m and 48-90 m respectively.

TABLE 4

GAMMA RADIONUCLIDE ACTIVITY (BECQUERELS/KG) IN LAKE ONTARIO BASINS*

Mid-Depths	Toronto (90 m)	Darlington (80 m)	Port Cobourg (48 m)
^{134}Cs	0.0	0.0	0.0
^{60}Co	0.0	0.0	0.0
^{137}Cs	10.36	6.66	4.44
Lake Basins	**Niagara Basin (115 m)**	**Mississauga Basin (180 m)**	**Rochester Basin (218 m)**
^{134}Cs	0.0	0.0	0.0
^{60}Co	1.22	0.78	2.55
^{137}Cs	358.86	307.07	292.27

* Lake basins are those classified by Thomas et al. (1972).

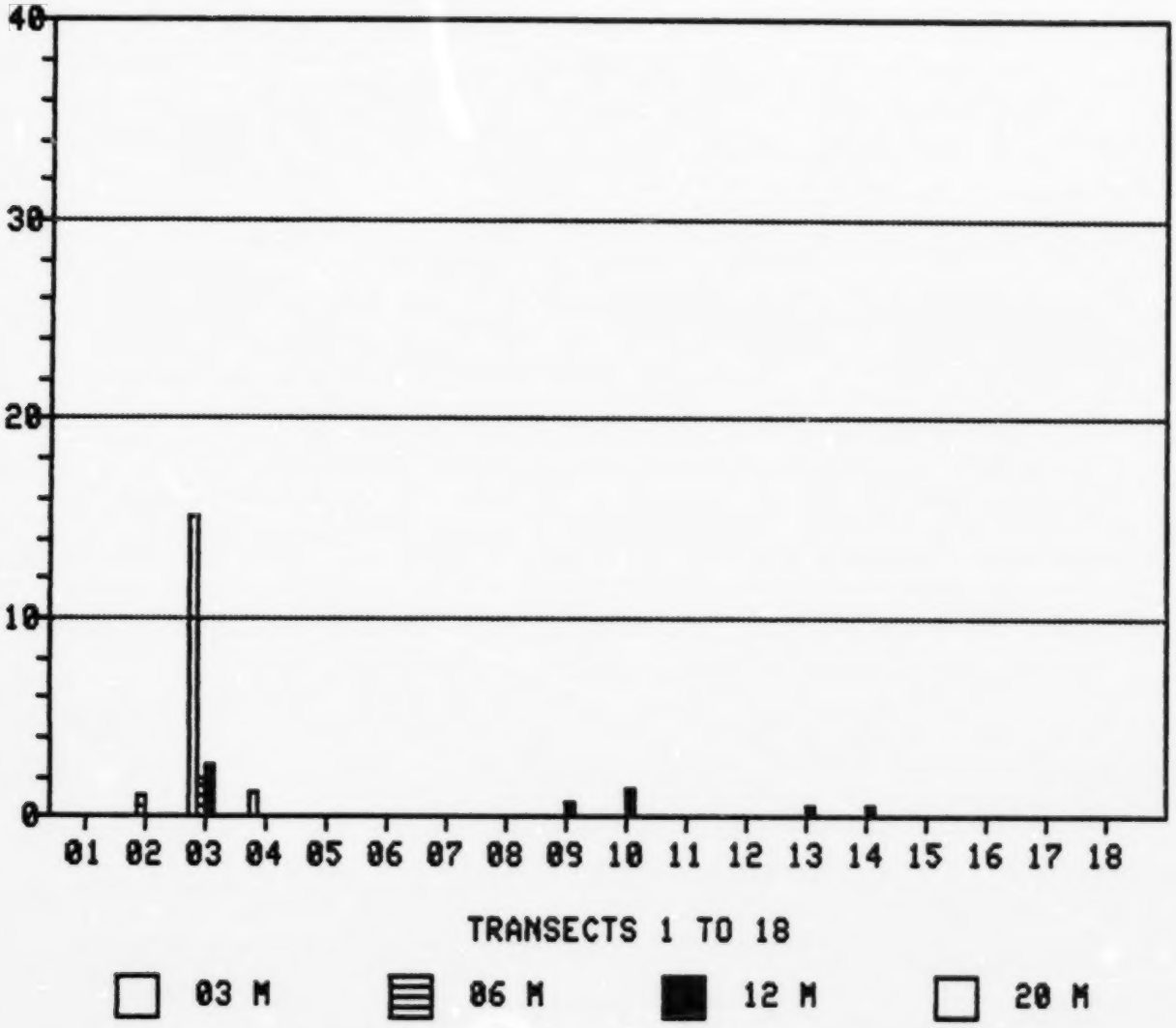

FIGURE 3

Distribution of Cobalt-60 In The Nearshore Sediments (3,6,12 and 20 M Depths) from Pickering to Darlington. Transects Initiated from 4 km West of Pickering 'A' NGS Discharge (Transect 3) to 4 km East of Darlington GS (Transect 16).

DISCUSSION

Eleven gamma emitting radionuclides were observed in Lake Ontario sediments. Radionuclides attributable to background and weapons testing were strongly inter-correlated, and their distributions were well represented by total gamma activity, which was primarily associated with percent organic material in the sediments. Three radionuclides were potentially of CANDU origin, however only ^{60}Co showed a distributional association with the Pickering 'A' NGS CCW discharge. Neither ^{134}Cs nor the ubiquitous ^{137}Cs in the present study were source related.

^{60}Co was adsorbed to the sediments primarily within a 2 km radius of Pickering 'A' NGS. While the adsorption mechanism is not clearly understood, several factors are known to influence the process. The quantity of organic matter has been suggested by Revsin et al, (1984) as a good indicator of potential sites for ^{60}Co accumulation. Ophel and Fraser (1973) however, did not find any significant difference in ^{60}Co concentrations between organically rich and poor sediments (55% and 2% dry weight respectively). Mahara and Kudo (1981), found that ^{60}Co was fixed on a sand substrate under aerobic conditions and a pH of 6.0 to 8.0. Similar conditions (aerobic, pH 7.2-7.8) would have existed in the immediate vicinity of the station's discharge during sample collection. These conditions may have resulted in preferential adsorption of ^{60}Co to sediments in the immediate vicinity of the Pickering 'A' NGS discharge.

Additional locations showing ^{60}Co activity included sites 12-14 km and 20-22 km east of the discharge, as well as the lake basins. The origin of ^{60}Co at sites east of the discharge is not clear, since no association with a known source was apparent. The lake basins are sites of deposition, characterized by silty clay sediments. Many contaminates present in the lake basins, including PCB, mirex, mercury, chormium, vanadium, and arsenic have been shown to primarily originate from the Niagara River drainage system (Thomas, 1983). Given the depositional characteristics of this area, and the absence of ^{60}Co from samples at the 48-90 m depths along the northshore, ^{60}Co present in the lake basins therefore may have originated from the Niagara River drainage system.

^{137}Cs activity in the vicinity of Pickering and Darlington was well described by a regression relationship with percent organic matter and depth. The lack of fit observed from two deep water sites, suggests activities present in the basins are strongly influenced by additional variables. A single occurrence along transect 7 was higher than expected, however no association with a point source was apparent. ^{137}Cs activity near the discharge

was typical for this shoreline. The relative contribution from Pickering 'A' NGS and from weapons testing to ^{137}Cs concentration in the sediments could not be determined.

It is interesting to note that sediments do not simply act as a sink for gamma radionuclides but serve more as a reservoir, and may release radionuclides under certain conditions. Mahara and Kudo (1981) have shown that ^{60}Co may be released from a sand sediment under anaerobic conditions. Stanners and Aston (1982) have shown that radiocesium nuclides can be desorbed up to 30% of their initial activity, indicating at least a partially reversible process. The adsorption/desorption behavior is also dependent upon the chemical species of the radionuclide released (OHRD Report 82-271-K).

In summary, therefore, analysis of Lake Ontario sediments indicated that the only detectable gamma radionuclide related to CANDU operations was ^{60}Co. The fate of the radionuclide appeared to be sediment adsorption in the immediate vicinity of the discharge. It is suggested that based on the lakewide depositional patterns for several (pollutants) or substances, that the offshore occurrence of ^{60}Co may have originated from the Niagara River watershed.

CONCLUSIONS

1) Eleven detectable gamma emitting radionuclides contributed to the total gamma activity observed from samples of Lake Ontario sediments.

2) Total gamma activity consisted largely of naturally occurring and weapons testing radionuclides, which were generally associated with percent organic matter in the sediments, and hence depth.

3) ^{137}Cs occurred in all samples collected throughout Lake Ontario, and was strongly correlated with background and weapons testing radionuclides. The distribution of ^{137}Cs in the vicinity of Pickering and Darlington was well described by the relationship with percent organic matter and depth.

4) Radionuclides which were the most probably indicators of recent CANDU activity (^{60}Co, ^{134}Cs) accounted for less than 1% of the total gamma activity, and were poorly correlated with other radionuclides.

5) Of the three potential CANDU released radionuclides (^{60}Co, ^{134}Cs, ^{137}Cs) only ^{60}Co probably originated from Pickering 'A'

NGS. Only distance from the discharge and depth appeared to influence the distribution of these occurrences.

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